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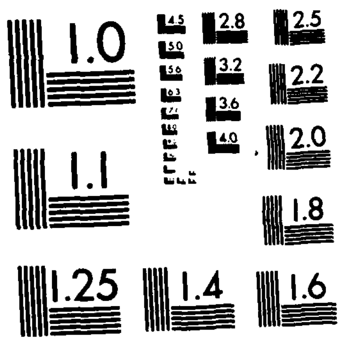

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NRL Report 8761

Dependence of Thermoluminescence Output on Temperature During Irradiation for Several Thermoluminescence Phosphors

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Several thermoluminescence phosphors were irradiated at elevated temperatures to determine if the thermoluminescence output was increased. Of the phosphors tested, only LiF:Mg showed any significant increase. Part of this increase can be attributed to annealing effects in the phosphor before and after exposure, while the remainder appears to be intrinsic to the phosphor temperature during the deposition of energy.

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CONTENTS

DEPENDENCE OF THERMOLUMINESCENCE OUTPUT ON TEMPERATURE DURING IRRADIATION FOR SEVERAL THERMOLUMINESCENCE PHOSPHORS

INTRODUCTION

Several investigators have reported an increased thermoluminescence (TL) output for dosimeters irradiated at elevated temperatures (~40° to 150°C) relative to those irradiated at room temperature. Gorbics et al.[1] observed a 5 to 10% increase in peak height for CaF_2:Mn dosimeters irradiated for 7 min at 100° to 150°C vs those irradiated at room temperature and below. Burke[2] reported an increased response (peak height) of 5 to 10% for CaF_2:Mn dosimeters exposed to low dose rates (~ 5 μR/h) for 30 days at temperatures of 52° and 60°C, while Tobias[3] reported a 5 to 10% increase in glow-curve area for LiF(TLD 100,700) irradiated for 1 h at 50° to 100°C. More recently, Sunta et al.[4] have reported a 20% increase in peak height for rapidly cooled LiF(TLD-100) dosimeters irradiated for 1 min at 80° to 120°C.

Exposure for any period of time at an elevated temperature may be considered to be a series of exposures with a predose anneal at the elevated temperature, an irradiation at the elevated temperature, followed by a postdose anneal at the elevated temperature. During the irradiation period, the percentage of predose annealing time increases from 0 to 100%, while the postdose annealing time decreases from 100 to 0%. Glow-peak growth resulting from annealing after exposure was first reported by Lucas[5] and later by other investigators[6-12]. Booth et al.[10] and Johnson[12] also observed a glow-peak growth up to 40% for rapidly cooled LiF:Mg(TLD-700) annealed at 22° to 100°C *before* exposure. Hence, for rapidly cooled LiF:Mg, increased TL output (glow-peak) for dosimeters irradiated at elevated temperatures can be explained by annealing effects in the phosphors, rather than through some mechanism related directly to the temperature during the actual deposition of energy in the phosphor during irradiation.

Kos et al.[13], citing optical absorption measurements, suggested that the results of Sunta et al.[4] could be explained by the thermal conversion of trapping centers ($Z_2 + e^- \rightarrow Z_2$) during irradiation at elevated temperatures. During subsequent readout, the Z_2 centers ($Mg^{2+} - F^1$ pairs) are converted to Z_3 centers (Mg^{2+} - F pairs) by the liberation of electrons causing the principal TL peak in LiF:Mg. Kos et al. do not state whether the $Z_2 + e^- \rightarrow Z_2$ conversion occurs only with irradiation during annealing, or is enhanced—or reduced—by irradiation during annealing. The optical absorption results of Jackson and Harris[8] for dosimeters annealed for 1 h at 80°C *prior* to irradiation at room temperature are strikingly similar to those of Kos et al. for dosimeters irradiated during annealing at 100°C. Thus the significance of irradiation *during* annealing on optical absorption is obscure. Based on their optical absorption measurements, Jackson and Harris[8] postulated trap migration and aggregation to explain peak-height growth for dosimeters annealed at elevated temperatures *before* irradiation. Citing those studies, Booth et al.[10] and Johnson[12] postulated trap migration and aggregation to explain peak-height growth for dosimeters annealed *before* or *after* irradiation.

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Tobias[3] also suggested that the increase he observed in TL area could be explained by the trap aggregation theory of Jackson and Harris. However, neither Booth et al. nor Johnson observed any increase in TL area for LiF(TLD-700) annealed at elevated temperatures before or after exposure.

Makajima[14] has proposed a TL model that assumes that an exothermic effect occurs along the path of the charged particles released during irradiation and that the temperature of the phosphor is added to this effect, thus increasing the probability of a charge carrier being trapped. Based on this model, one would expect an increase in TL output independent of annealing effects in the phosphor. Also, considering the "track interaction model" of LiF[15], one might expect an increased probability of charge carriers being trapped due to increased migration range at elevated temperatures. We therefore performed a series of experiments in an attempt to determine the increased TL output (if any) of dosimeters irradiated at elevated temperatures, and whether this increase was caused by changes in the phosphors induced by annealing or by some mechanism directly related to the temperature during the irradiation.

EXPERIMENTAL PROCEDURE

The phosphors used in this study were: LiF:Mg(TLD-700), CaF_2:Mn, CaF_2:Dy, CaF_2:Tm, and $Li_2B_4O_7$:Mn. All were in the form of blocks, 3.2 mm × 3.2 mm × 0.9 mm thick, and were obtained from the Harshaw Chemical Company, Solon, Ohio. Before irradiation, all dosimeters were heated at 400°C for 1 h and cooled to room temperature in a nonlinear, but reproducible fashion in approximately 10 min. In addition, the LiF dosimeters were either cycled once in a Harshaw Model 2000 TL analyzer using a linear readout schedule of 5°C/s to 300°C for a total readout time of 60 s, or annealed overnight (16 h) at 80°C. This latter "standard annealing" procedure[16] is used to reduce the low-temperature TL peaks, while the rapid cooling following readout in the reader enhances these peaks[10,12,16].

To determine the effect of irradiation temperature, dosimeters were dropped on a planchet held at temperatures from 20° to 300°C. The planchet was continuously irradiated using a Co-60 source. Irradiation time was 1 min giving a total exposure of approximately 100 mR. At the end of the irradiation, dosimeters were immediately removed from the planchet and cooled to room temperature in approximately 10 s. They were read approximately 1 h afterward by using the previously mentioned readout schedule of 5°C/s to 300°C. Since the low-temperature peak at 110°C interferes with the determination of the main peak-height in CaF_2:Dy, these dosimeters were given a prereadout anneal for 15 min at 100°C. None of the other phosphors received any prereadout anneal. Glow-curve area and peak-height(s) were simultaneously recorded for each dosimeter. After readout, all dosimeters were given an identical calibration exposure at 20°C to allow corrections for differences in TL sensitivity of the individual dosimeters and to allow all dosimeter readings to be normalized to their response at 20°C. All experiments were repeated at least twice, with a minimum of four dosimeters being irradiated at each temperature.

RESULTS AND DISCUSSION

Lithium Fluoride

Figure 1 shows the results for LiF(TLD-700) dosimeters which were unannealed except for being cycled once through the reader prior to irradiation. The increase in peak-height at 120°C is greater (∼20%) that that reported by Sunta et al.[4] for rapidly cooled dosimeters. This is perhaps as expected since Sunta et al.[4] allowed their dosimeters to remain on the planchet during cooling after exposure, which would cause fading due to emptying of traps. Also, their dosimeters may have

been cooled more slowly during the initial annealing, resulting in fewer low-temperature traps available for conversion to high-temperature traps, the mechanism postulated by Booth et al.[10] and Johnson[12] to explain glow-peak growth during annealing. Their dosimeters may have also come from a different batch which had fewer low-temperature traps. We have noted a batch variation of up to a factor of two in the magnitude of the low-temperature peaks of dosimeters purchased during the past 10 years. Shown in Fig. 2 are glow curves of dosimeters used for most of the present study (A) and those having considerably fewer low-temperature traps (B). Dosimeters from batch (B) showed less than half the glow-peak increase shown in Fig. 1. The increased peak-height shown in Fig. 1 is also greater than the maximum peak-height growth ($\sim$30%) observed by Johnson[12] for LiF(TLD-700) dosimeter blocks having the same number of low-temperature traps which were given predose or postdose anneals for *longer* times at *lower* temperatures. Note, however, that there is no increase in glow-curve area at elevated annealing temperatures. Based on these results, and the previously cited references showing glow-peak growth during predose and postdose annealing, it might seem reasonable to attribute this glow-peak increase to annealing effects in the phosphor rather than to some mechanism intimately related to the temperature during irradiation as was done by Sunta et al.[4]. However, of the references cited, only the studies of Lucas[5] were done using annealing times as short as those of Sunta et al. and the present study. Hence, they may not be directly applicable.

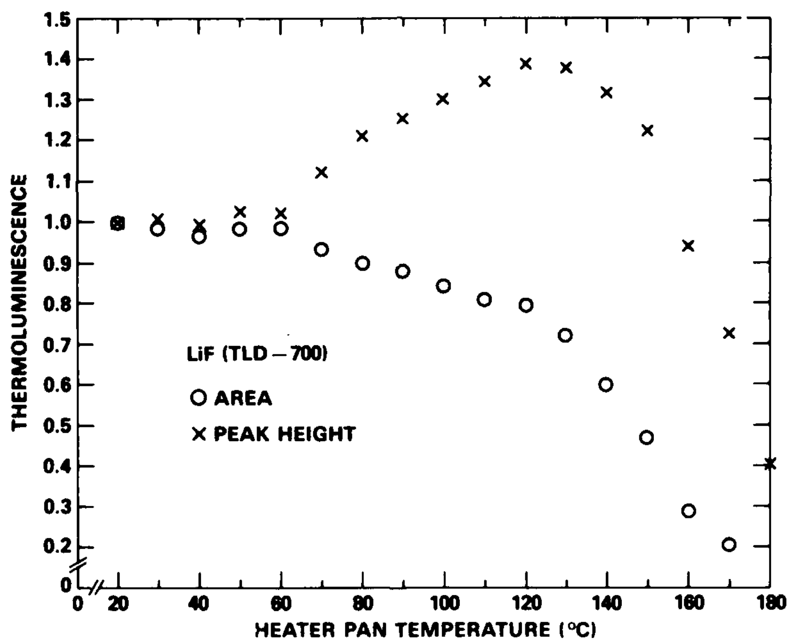

Fig. 1 — Response of LiF(TLD-7000) thermoluminescence dosimeters as a function of heater pan temperature during irradiation relative to the same dosimeters given a calibration exposure at 20°C. Dosimeters were cycled once through the TL reader immediately prior to exposure, and were then rapidly cooled.

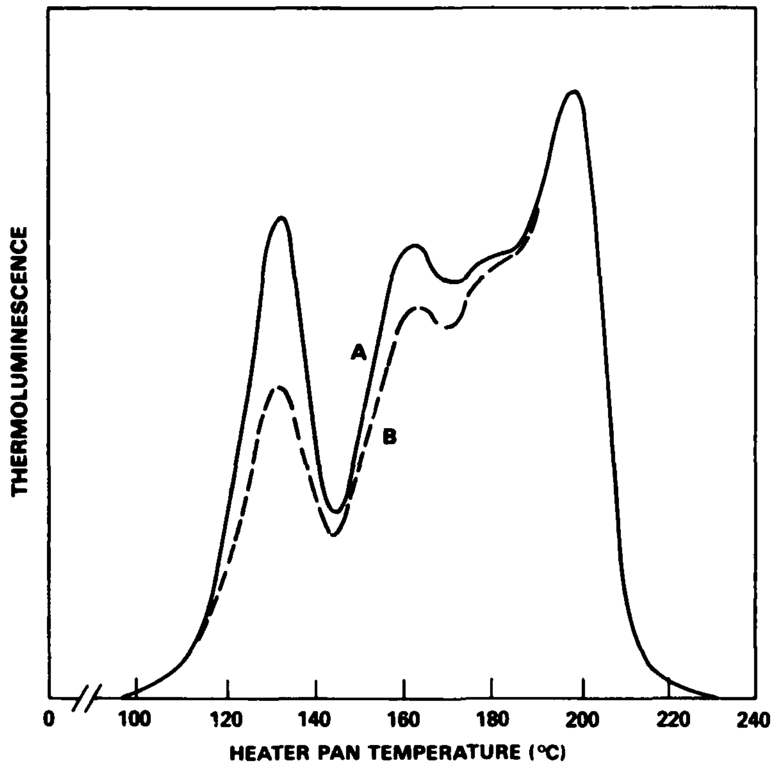

Fig. 2 — Glow curves, thermoluminescence vs heater pan temperature, for LiF(TLD-700) dosimeters used for most of this study (A) and for dosimeters having fewer low-temperature peaks (B). Dosimeters were cycled once through the TL reader immediately prior to exposure and read 20 min later.

Shown in Fig. 3 are the results for LiF(TLD-700) dosimeters given the "standard annealing" of 16 h at 80°C prior to exposure. Here we see a 20% increase in *both* peak height and glow-curve area, indicating that elevated temperature during irradiation increases TL output even when few low-temperature peaks are present. Dosimeters from batch (B) showed an 8% increase in peak height and area, indicating a correlation between the number of low-temperature traps and TL increase, even when the low-temperature traps are drastically reduced by the "standard annealing" procedure. The increase in glow-curve area and peak height, shown in Fig. 3, suggests that some mechanism other than trap conversion during annealing may be causing some of the increased TL peak height observed in Fig. 1 with the rapidly cooled "unannealed" dosimeters.

In an attempt to rule out annealing effects as the cause of the increased response shown in Fig. 3, an experiment was performed in which "standard annealed" dosimeters were dropped on the heated planchet for 1 min, removed and cooled to room temperature, dosed to 5 R using a Co-60 source, then read. Other dosimeters were dosed to 5 R, dropped on the heated planchet for 1 min, removed and cooled to room temperature, then read. A third group was dropped on the heated planchet for 30 s, removed and cooled to room temperature, dosed to 5 R, again heated for 30 s on the planchet, removed and cooled to room temperature and read. Figure 4 shows the results of this experiment. None of the annealing procedures, in which the dosimeters were dosed at room temperature, caused an increase in peak height for any of the dosimeters, and only dosimeters annealed prior to dosing showed an increase in glow-curve area. This increase is caused by the growth in the low-temperature glow peaks [16], which are faded by the postdose anneal of the other annealing

cycles. Thus it would appear that annealing effects are not the principal cause of the increase in peak height and area shown in Fig. 3; but rather is some mechanism involved that is intimately related to the temperature during irradiation.

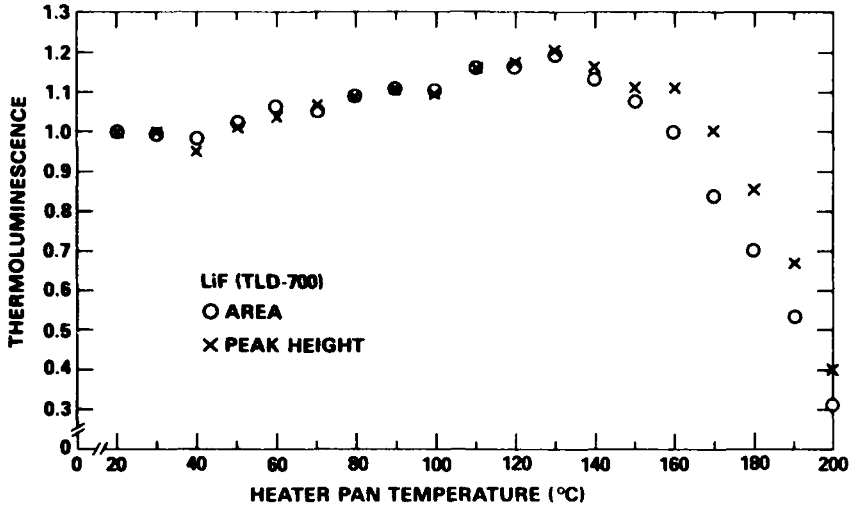

Fig. 3 — Response of LiF(TLD-700) thermoluminescence dosimeters as a function of heater pan temperature during irradiation relative to the same dosimeters given a calibration exposure at 20°C. Dosimeters were given a low-temperature anneal at 80°C for 16 h prior to exposure.

To confirm that the peak height increase shown in Fig. 1 was not solely due to annealing processes, the previously described experiment was repeated using rapidly cooled dosimeters. Peak height increase was 15% for dosimeters dosed before or at the middle of the annealing period. Dosimeters dosed after annealing showed less than 10% increase in peak height. None of the dosimeters showed an increase in glow-curve area. This experiment indicates that less than half of the peak height increase shown in Fig. 1 is caused by annealing effects, presumably the conversion of low-temperature traps to high-temperature traps. Note that it is not possible to unequivocally separate annealing effects from effects caused by elevated temperature during irradiation, since the temperature of the phosphor cannot be elevated without annealing the phosphor.

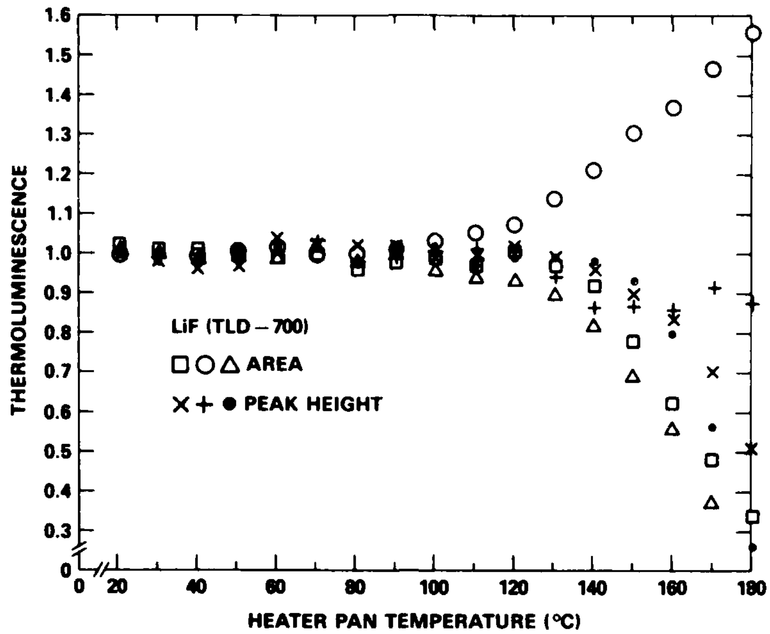

Fig. 4 — Response of LiF(TLD-700) thermoluminescence dosimeters vs annealing temperatures for: dosimeters annealed for 1 min after exposure at 22°C (▫, x); dosimeters annealed for 1 min prior to exposure at 22°C (0, E); and dosimeters annealed for 30 s before and after exposure at 22°C (▵, •). All dosimeters were given a low-temperature anneal at 80°C for 16 h prior to the experiment.

The Other Phosphors

Figure 5 shows the results for CaF_2:Mn dosimeters. Contrary to earlier referenced studies[2,6[, there is no evidence of any increase in peak height or area, suggesting that annealing changes may have been responsible for the previously reported results in which exposures were given over a longer period of time.

Figure 6 shows the results for CaF_2:Dy. There is no evidence of any increased response at elevated temperatures.

Figure 7 shows the results for CaF_2:Tm. Although there is more statistical error in these results, it appears that there is about a 5% increase in the height of the 240°C peak, with no increase in the height of the 150°C peak or in the total glow-curve area. Since we felt that the increase in the 240°C peak height might be caused by annealing effect, e.g., trap conversion of the lower temperature traps as in LiF, we performed experiments in which dosimeters were annealed for 1 min at 120°C before and after exposure, and for 30 min at 100°C before and after exposure. We found no evidence of increased TL output.

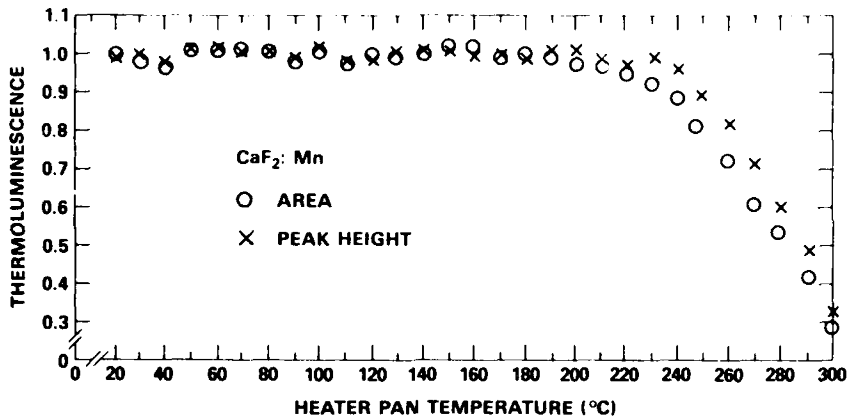

Fig. 5 — Response of CaF_2:Mn thermoluminescence dosimeters are a function of heater pan temperature during irradiation

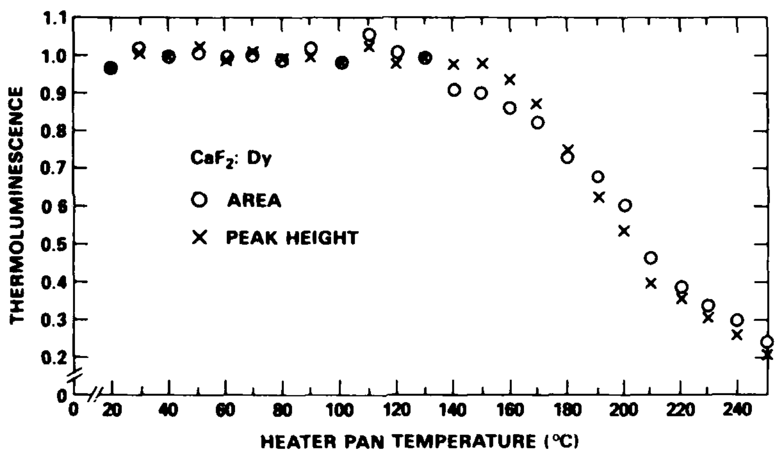

Fig 6 — Response of CaF_2:Dy thermoluminescence dosimeters as a function of temperature during irradiation

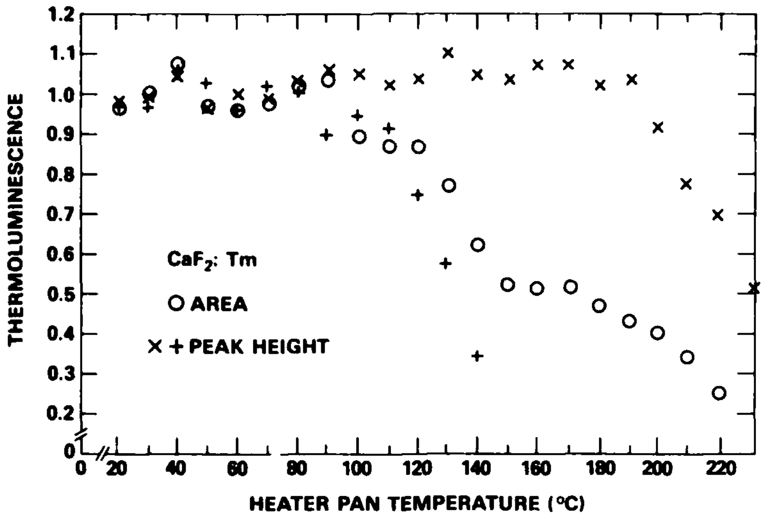

Fig. 7 — Response of CaF_2:Tm thermoluminescence dosimeters as a function of temperature during irradiation (+, 150°C peak height; x, 240°C peak height)

Figure 8 shows the results for $Li_2B_4O_7$:Mn dosimeters. Because of the low light output, we were only able to obtain data for glow-curve area.

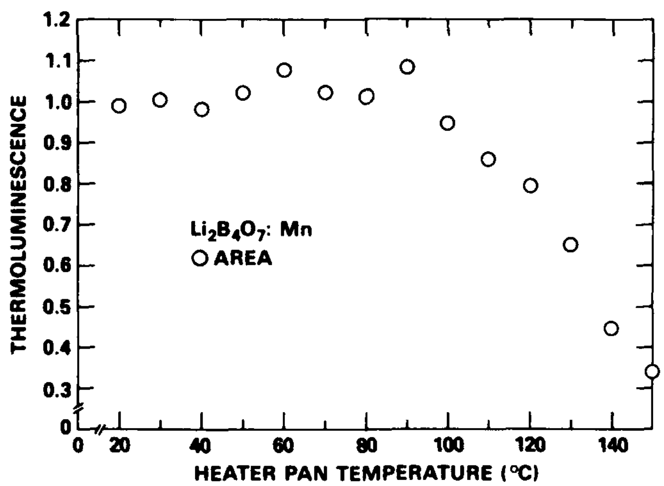

Fig. 8 — Response of $Li_2B_4O_7$:Mn thermoluminescence dosimeters as a function of temperature during irradiation

CONCLUSIONS

Of the phosphors tested, only LiF:Mg(TLD-700) exhibits any significant increase in TL output for dosimeters irradiated at elevated temperatures. For rapidly cooled dosimeters, some of this increase is probably caused by annealing effects before and after exposure as has been previously postulated to explain glow-peak growth in dosimeters given predose and postdose

anneals[10,12,13]. Our results for LiF:Mg dosimeters given "standard annealing" of 400°C for 1 h + 80°C for 16 h indicate that peak height and flow curve area increase is also caused by some process intimately related to the temperature during irradiation. Our studies indicate a correlation with the number of low-temperature traps for both processes. Any successful theory of thermoluminescence for LiF:Mg must be able to account for both of these phenomena. The correlation of TL increase with the number of low-temperature traps for LiF:Mg and the absence of significant TL increase for the other phosphors when irradiated at elevated temperatures indicates that the "exothermic effect" theory proposed by Nakajima[14] is not generally applicable. Further studies are planned to elucidate the mechanisms involved in TL increases.

When using these TL materials at elevated temperatures, caution should be exercised to ensure that TL fading and/or growth do not introduce errors in the results obtained.

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